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Observations of the first aerosol indirect effect in shallow cumuli

Larry K. Berg,[1] Carl M. Berkowitz,[1] James C. Barnard,[1] Gunnar Senum,[2] and Stephen R. Springston[2]

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[1] Data from the Cumulus Humilis Aerosol Processing Study (CHAPS) are used to estimate the impact of both aerosol indirect effects and cloud dynamics on the microphysical and optical properties of shallow cumuli observed in the vicinity of Oklahoma City, Oklahoma. Not surprisingly, we find that the amount of light scattered by clouds is dominated by their liquid water content (LWC), which in turn is driven by buoyancy and cloud dynamics. However, removing the effect of cloud dynamics by examining the scattering normalized by LWC shows a statistically significant sensitivity of scattering to pollutant loading (increasing at a rate of 0.002 m^2 g^{-1} $ppbv^{-1}$). These results suggest that even moderately sized cities, like Oklahoma City, can have a measureable impact on the optical properties of shallow cumuli. **Citation:** Berg, L. K., C. M. Berkowitz, J. C. Barnard, G. Senum, and S. R. Springston (2011), Observations of the first aerosol indirect effect in shallow cumuli, *Geophys. Res. Lett.*, *38*, L03809, doi:10.1029/2010GL046047.

1. Introduction

[2] *Twomey* [1977] postulated that increasing the number of particles available to act as cloud condensation nuclei (CCN) while holding the liquid water content (LWC) constant inside a cloudy column of air would lead to an increase in the cloud drop number concentration (CDNC), a decrease in the droplet effective radius (r_{eff}, defined as the ratio of the third and second moments of the drop size distribution) and an increase in the cloud albedo. This phenomenon is called the Twomey effect or First Aerosol Indirect Effect (FAIE). Changes in the supersaturation associated with variation of the cloud updraft velocity also influence r_{eff}. In addition, the impact of the FAIE on cloud brightness and precipitation efficiency can be tempered by changes in the relative dispersion of the cloud drop size distribution (defined as the ratio of the standard deviation of the droplet diameter to the mean droplet diameter) [e.g., *Liu and Daum*, 2002]. Coincident measurements of cloud microphysical properties, cloud dynamics, particle loading, and pollution levels are an optimal way to document the FAIE, and to evaluate the relative influence of aerosols and cloud dynamics on cloud optical properties.

[3] Evidence of the FAIE in continental and maritime stratiform clouds, as well as in cumuli found in the vicinity of large urban areas has been presented in the literature. Changes in the microphysical properties of warm stratocumulus over the oceans or land have been documented in a number of studies [e.g., *Radke et al.*, 1989; *Durkee et al.*, 2000; *McFarquhar and Heymsfield*, 2001; *Feingold et al.*, 2003; *Twohy et al.*, 2005]. Changes in the microphysical structure of cumuli downwind of St. Louis, Missouri were documented by *Fitzgerald and Spyers-Duran* [1973], while *Alkezweeny et al.* [1993] presented evidence of changes in stratocumuli downwind of Denver Colorado. More recently, attention has been focused on details of aerosol-cloud interactions in shallow cumuli in the vicinity of Houston, Texas [*Lu et al.*, 2008].

[4] While evidence of the FAIE has been identified in several major metropolitan areas (populations of several million individuals), few studies have looked for the effects in any of the smaller cities of North America (populations on the order of hundreds of thousands of individuals), which are often the only significant source of pollution for hundreds of square miles. In the study presented here, evidence of the FAIE will be presented for an ensemble of shallow cumuli observed near Oklahoma City, Oklahoma (population of approximately 540,000; http://quickfacts.census.gov/qfd/states/40/4055000.html), which is representative of many smaller cities throughout North America.

2. Data and Methods

[5] The Cumulus Humilis Aerosol Processing Study (CHAPS) was conducted during June 2007 and was designed to investigate cloud-aerosol interactions in the vicinity of Oklahoma City, Oklahoma [*Berg et al.*, 2009]. The primary instrument platform was the U.S. Department of Energy's Gulfstream 1 (G-1) aircraft, which was configured to make *in situ* measurements of the chemical and optical properties of aerosols, cloud microphysics, trace gas concentrations, and meteorological variables. The analysis to be presented here will make use of a subset of these observations. A Droplet Measurement Technology Cloud Aerosol and Precipitation Spectrometer (CAPS) probe configured to use 20 unequal-sized bins to measure particles ranging in diameter from 0.63 to 50 μm provided the droplet size distribution. The CAPS measurements were used to determine the CDNC, LWC, r_{eff} and the relative droplet dispersion. CO was measured using a vacuum UV fluorimeter and a gust probe integrated into the nose of the G-1 to measure the vertical velocity (w). The flight pattern used during CHAPS included overlaid straight-line transects flown below-, within-, and above the cloud layer. The analysis to follow makes use of data collected on six separate days when shallow cumuli were present and includes data from 28 flight legs and a total of 768 individual cloud penetrations, which were generally flown near cloud base as reported in flight logs. As would be expected when sampling cumuli of limited

[1]Pacific Northwest National Laboratory, Richland, Washington, USA.
[2]Brookhaven National Laboratory, Upton, New York, USA.

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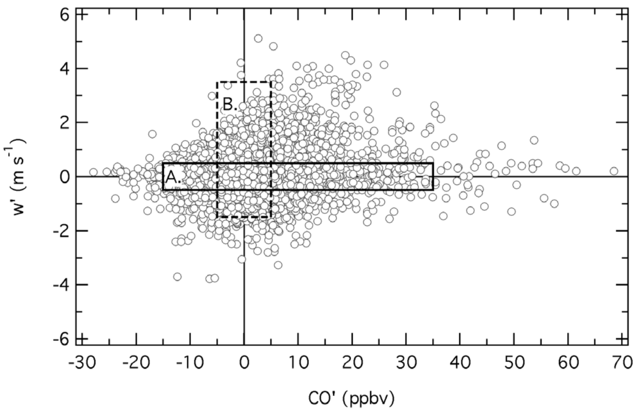

Figure 1. Scatter plot of all 1-second averages of CO' and w' measured during cloud penetrations made during CHAPS. Points within Box A were used to examine the sensitivity of cloud parameters to variations in pollutant loading for small variations in updraft strength. Points in Box B were used to examine the sensitivity of cloud parameters to variations in updraft strength for small variations in pollutant loading.

horizontal extent with an aircraft flying at approximately 100 ms^{-1}, the time spent within a given cloud was typically short, averaging between 4 and 5 seconds.

[6] A number of different parameters could be used to indentify air parcels that have been influenced by emissions associated with Oklahoma City before being lofted into a cloud. During CHAPS, the urban plume was generally marked by an increase in accumulation mode particles (defined to include particles 0.5 to 2 μm in diameter) as well as an increase in the mixing ratio of CO. Previous studies have used CO as a tracer of urban emissions [e.g., *Kleinman et al.*, 2008] because it has the advantage over many other tracers of being conserved with passage through clouds. As a result, the relative pollutant loading can be determined for each individual cloud. In contrast, *Feingold et al.* [2003] and *Lu et al.* [2008] used the number of subcloud accumulation mode particles to define clean and dirty conditions. The following analysis assumes the anthropogenic particle loading encountered by the cloud droplets is proportional to the local concentration of CO.

[7] Background mixing ratios of CO measured upwind of Oklahoma City during CHAPS ranged from 100 to 140 ppbv. In order to focus the analysis on the impact of fresh emissions the mean and linear trend of CO computed for each individual flight leg were removed from the 1-second observations of CO, yielding a perturbation CO value (CO'). Thus values of CO' near zero are representative of regional values found outside of the Oklahoma City plume.

[8] A similar treatment was applied to the time series of w but for different reasons. The mean w measured by aircraft mounted gust probes is generally considered to be unreliable and is assumed to be 0 ms^{-1}. Therefore, the mean and linear trend of w was computed for each leg of the flight pattern and was removed from the observed w, yielding a perturbation measure of w (w'). This treatment of w is typical of studies of turbulence where the fluctuations of a variable are of greater interest than the mean quantity of the variable itself.

[9] Values of CO' and w' measured during all CHAPS cloud penetrations are shown in Figure 1. A preliminary examination of the CHAPS data suggested that CDNC was a function of both the pollutant loading and cloud dynamics. To isolate these effects, results in this study focus on average values computed for points lying in the two boxes highlighted in Figure 1. Box A is defined by values of CO' between −15 ppbv and 35 ppbv, and w' between −0.5 and 0.5 ms^{-1}, with points in this box used to examine the sensitivity of cloud droplets to pollutant loading for a relatively small range of w'. Box B is defined by values of CO' between −5 and 5 ppbv, and w' between −1.5 and 3.5 ms^{-1}. Points in this box will be used to examine the sensitivity of cloud droplets to updraft strength for a relatively small range of CO$'$. The two boxes shown in Figure 1 represent approximately 80% of the observations. Even so, bins with less than 10 observations have been excluded from the analysis. Average values of CDNC, LWC, r_{eff}, and dispersion were determined for equally spaced bins of CO' and w' defined to be within boxes A and B.

3. Results and Discussion

[10] The results of this analysis provide evidence that FAIE can affect cumuli downwind of moderately sized cities. The nonparameteric Wilcoxon Rank test was used to confirm the statistical significance (at the 0.05 level) of the trends of all of the variables reported here (CDNC, r_{eff}, relative dispersion, and the two variables related to the optical scattering). The CDNC is found to increase with both CO' and w' (Figures 2a and 2d). Increasing w' from −1 to 3 m s^{-1} for cases in which values of CO' are between −5 and 5 ppbv (points found in Box B shown in Figure 1), leads to an increase of nearly a factor of 1.8 in the CDNC. The CDNC increases by a factor of 1.7 with increasing CO' as can be seen for points for which CO' ranges from −10 to 30 ppbv and w' are between −0.5 and 0.5 ms^{-1} (points found in Box A shown in Figure 1). As suggested by *Twomey* [1977], these changes in the CDNC could lead to greater cloud optical thicknesses, assuming that the LWC of the cloud is constant. No systematic change in LWC was associated with changes in pollutant loading.

[11] The effective radius, r_{eff}, is found to change with both w' and CO' (Figures 2b and 2e). The increase in r_{eff} for both updrafts and downdrafts is associated with changes in the underlying cloud drop size distribution (Figure 3). Relative to measurements associated with values of w' near 0 ms^{-1}, measurements associated with large values of w' ($w' \sim 3$ ms^{-1}), show a large increase in bigger drops that in turn leads to an increase in r_{eff}. In downdrafts ($w' \sim -1$ ms^{-1}), there is a decrease, relative to when $w' \sim 0$, in the number of smaller drops, but in this case there is little change in the number of larger drops, leading to an increase in r_{eff} measured in downdrafts. This change may be attributed to drying in the downdrafts reducing the number of small drops. In cases when w' is approximately zero, the spectrum is shifted to the left, resulting in a relatively small value of r_{eff}. Consistent with *Twomey*'s [1977] prediction, r_{eff} decreases as pollutant loading increases (Figure 2e).

[12] Changes in the relative dispersion of the cloud drop size distribution, were found to be sensitive to the updraft

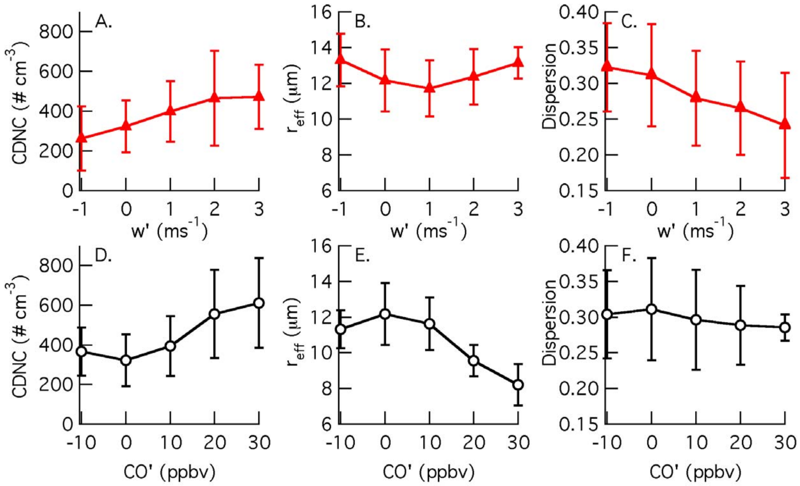

Figure 2. Mean value of (a) CDNC, (b) r_{eff}, (c) relative dispersion sorted according to bins of w' (with CO' ~ 0 ppbv; Box B in Figure 1) and (d) CDNC, (e) r_{eff}, and (f) relative dispersion sorted according pollutant loading (with $w' \sim 0$ ms^{-1}; Box A in Figure 1) measured during cloud penetrations. Error bars indicate the standard deviation of the observations.

strength but not the pollutant loading (Figures 2c and 2f). The dependence of the dispersion on the updraft strength has been documented previously [e.g., *Politovich*, 1993]. The relatively small sensitivity of the dispersion to pollutant loading at first seems to be at odds with past studies by *Martin et al*. [1994], *McFarquhar and Heymsfield* [2001, and *Liu and Daum* [2002], all of whom have reported larger values of the relative dispersion in polluted clouds. However, these earlier results were obtained in maritime conditions in which the clean clouds were considerably cleaner than the clean clouds observed during CHAPS (CO' < 0). For example, during the Indian Ocean Experiment (INDOEX) the condensation nuclei (CN) concentrations ranged from approximately 300 to 1500 cm^{-3} for clean to dirty conditions [*Heymsfield and McFarquhar*, 2001]. During CHAPS, observations of CN outside of the Oklahoma City plume were frequently greater than 2000 cm^{-3}. As suggested by *Liu and Daum* [2002], changes in the relative dispersion could counteract the FAIE. In the shallow cumuli studied here, however, the dispersion is not a strong function of the pollutant loading (Figure 2f), and does not counteract the FAIE.

[13] The LWC was determined by integrating the cloud drop size distribution measured by the CAPS probe and found to be strongly dependent on w' (not shown). The CAPS derived LWC was found to be consistent with the LWC measured using a Gerber PVM-100A probe that was also mounted on the G-1. The average LWC in downdrafts during CHAPS was approximately 0.4 g m^{-3}, while values of as large as 1.3 g m^{-3} were measured in the strongest updrafts.

[14] While the preceding results demonstrate the effect of pollution on microphysics they do not by themselves demonstrate a systematic change in the amount of light scattered by the clouds and, in turn, changes in the cloud optical depth. Light scattering by clouds was not measured during CHAPS. Instead, the volume scattering coefficient [*Goody and Yung*, 1989] (σ_s) was estimated using Mie theory [*Hansen and Travis*, 1974] and the CAPS derived cloud drop size distribution. While it has been reported [e.g., *Chýlek and Ramaswamy*, 1984; *Chýlek et al.*, 1996] that black carbon inside of cloud drops can have an impact on the absorption of light by clouds, given that the amount of black carbon over central Oklahoma is small [e.g., *Sheridan et al.*, 2001], and the impact of the aerosol on the cloud absorption is small, our calculations assumed that the mass of the CCN inside the drop had no effect on the cloud optical properties. Other studies, such as *Cess et al.* [1995], also suggest that aerosols do not have a strong impact on the cloud absorption. These calculations showed that to the first order, light scattering was dominated by changes in the LWC, increasing by nearly a factor of 5 as the LWC varies from 0.4 g m^{-3} to 1.3 g m^{-3} (not shown). The LWC is highly correlated with vertical velocity, and as anticipated, the light scattering by cloud drops increases with increasing w' (Figure 4a). Based on the changes in r_{eff}, one would expect that the total scattering would increase with

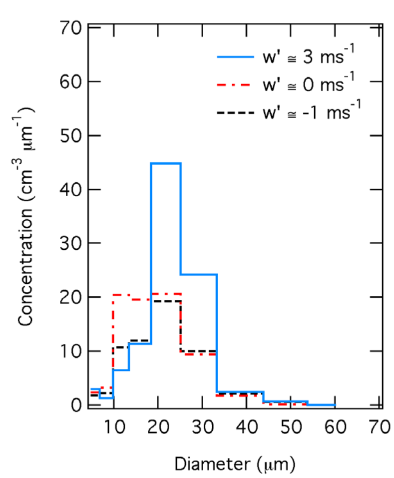

Figure 3. Average cloud droplet size distribution for binned values of CO' between −5 and 5 ppbv, and $w' \sim 3$ (blue), 0 (red), and −1 m s^{-1}.

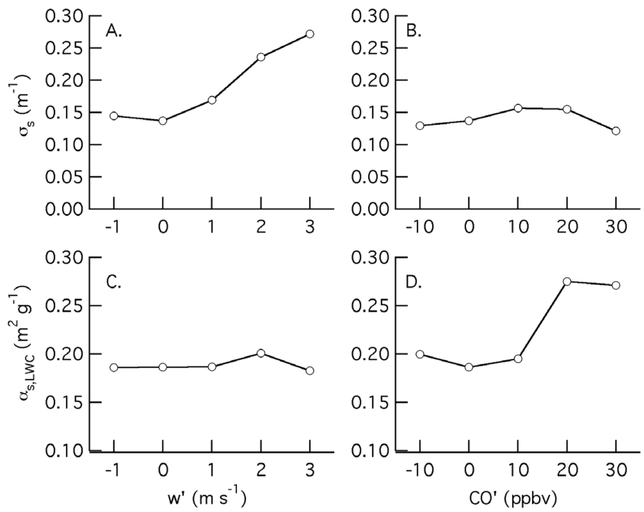

Figure 4. Mean values of σ_s sorted according to (a) bins of w' (with CO' ~ 0 ppbv; Box B in Figure 1) and (b) pollutant loading, (with $w' \sim 0$ ms^{-1}, Box A in Figure 1); and mean values of $\alpha_{s,LWC}$ sorted according to (c) bins of w' (with CO' ~ 0 ppbv; Box B in Figure 1) and (d) pollutant loading (with $w' \sim 0$ ms^{-1}, Box A in Figure 1).

increasing values of CO' (and values of w' near 0) as well. However, this was not observed, with the total scattering found to be nearly independent of CO' (Figure 4b). The total scattering of light by the clouds is strongly dependent on the LWC. While the LWC does not change in a systematic fashion with the variations in pollution, it is not constant. In order to account for these small variations in LWC, σ_s was normalized by the LWC, yielding a "normalized", or LWC scattering coefficient, ($\alpha_{s,LWC}$). In contrast to the σ_s, the $\alpha_{s,LWC}$ was found to be independent of w' (Figure 4c) and strongly dependent on pollutant loading (Figure 4d). Focusing only on the points highlighted in boxes A and B of Figure 1, it can be seen that when w' is small in magnitude, the normalized scattering increases at a rate of 0.002 m^2 g^{-1} ppbv^{-1} (Figure 4d). A Wilcoxon Rank test was used to confirm that the changes in $\alpha_{s,LWC}$ with pollutant loading are significant at the 0.05 level. In contrast, $\alpha_{s,LWC}$ is nearly independent of the w'. Although cloud microphysical properties are the result of many complex interactions involving cloud dynamics and aerosol properties, the use of $\alpha_{s,LWC}$ greatly reduces the dependence of scattering on cloud dynamics, with the sensitivity predicted by the FAIE illustrated in Figure 4d.

4. Conclusions

[15] Evidence of the FAIE in fields of shallow cumuli is presented using observations made during CHAPS. Statistically significant systematic changes in the CDNC, r_{eff}, and dispersion of the cloud drop size distribution are found to be a function of both the updraft strength and pollutant loading. While observations of the σ_s in clouds were not made during CHAPS, this quantity was computed using Mie Theory and the measured cloud drop size distributions. Values of σ_s were found to be strongly dependent on the LWC, which in turn is a strong function of w'. However, the $\alpha_{s,LWC}$ (i.e., the σ_s normalized by LWC) showed a much stronger, statistically significant, dependence on pollutant loading as measured by observations of CO. These results suggest that even moderately sized cities, like Oklahoma City, can have a measureable impact on the optical properties of shallow cumuli. While a myriad of factors influence the cloud microphysical properties, this work demonstrates the importance of considering both the cloud dynamics and the aerosol loading when investigating aerosol indirect effects.

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J. C. Barnard, L. K. Berg, and C. M. Berkowitz, Pacific Northwest National Laboratory, P.O. Box 999, Richland, WA 99352, USA.

G. Senum and S. R. Springston, Brookhaven National Laboratory, P.O. Box 5000, Upton, NY 11973, USA.